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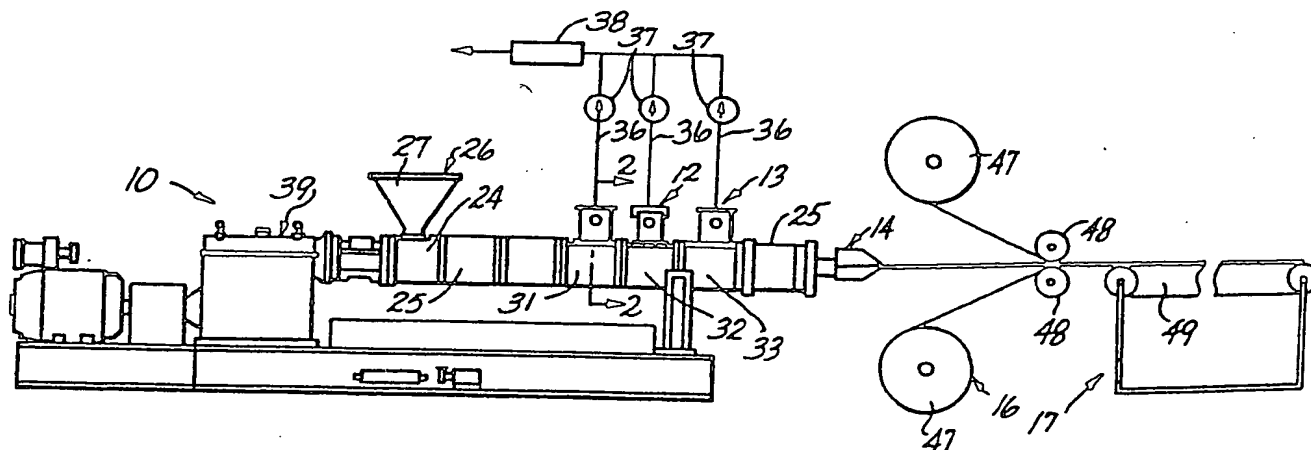
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(54) Title: HIGH PERFORMANCE PRESSURE SENSITIVE ADHESIVE TAPES AND PROCESS FOR MAKING THE SAME



(57) Abstract

A pressure sensitive adhesive tape comprises a carrier layer having a thickness of 0.25 to 2.0 millimeters and comprising an electron beam cured pressure sensitive adhesive matrix, 10 to 20 % by volume low density microspheres and at least one pigment in an amount sufficient to color the tape. Preferably, fumed silica is present in an amount up to 5 % by weight. The tape preferably has a skin layer on each side of the carrier layer. The skin layer has a coating thickness of 25 to 125 g/m² and comprises a pressure sensitive adhesive matrix free of rigid, low density microspheres. The process comprises first preparing an adhesive composition containing electron beam-curable adhesive polymer matrix, fillers and solvent. The composition is introduced and conveyed through a twin screw extruder (10). In the extruder, solvent is removed in one or more solvent removal units (11, 12, 13), and a solvent-free composition is extruded as the carrier layer. Skin layers may be coextruded with the carrier layer.

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10 HIGH PERFORMANCE PRESSURE SENSITIVE ADHESIVE TAPES
 AND PROCESS FOR MAKING THE SAME

Field of the Invention

15 This invention relates to pressure sensitive adhesive tapes and more particularly to electron beam-cured double-coated acrylic and rubber based pressure sensitive adhesive foam-like tapes and a process for making such tapes.

20 Background of the Invention

 A variety of double-coated foam tapes are being used for structural bonding in certain applications replacing spot welds, tack welds, or rivets. Such applications include, for example, the bonding of side molding to automobiles, fiberglass body panels to motor homes, plexiglass inspection windows onto equipment cabinets, and the like. The foam layer of these tapes usually has a polymer matrix based on polyethylene, polyurethane, polyvinyl chloride, or polychloroprene. These tapes exhibit poor conformability around curved substrates.

 U.S. Patent No. 4,223,067 to Levens, assigned to Minnesota Mining and Manufacturing Co., describes a method for making conformable foam-like acrylic pressure sensitive adhesive tapes using on web-polymerization

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1 technology. In the process, a mixture of monomers and
20 to 65 volume percent glass microbubbles is coated
onto a backing sheet and then polymerized to a pressure
sensitive adhesive state. The polymerization may be
5 initiated by ultraviolet light or less preferably by
heat if the mixture includes a heat-activatable
polymerization initiator.

The tapes disclosed by Levens are fairly elastic
under briefly applied stresses but exhibit low
10 elasticity under prolonged stress and therefore adhere
to rough and uneven surfaces. These tapes exhibit high
peel adhesion. The method of Levens, however, requires
a long duration to complete polymerization. This makes
the tapes expensive to produce. Moreover, coatings
15 having a thickness greater than about 0.2 mm involving
neat monomers tend to produce excessive bubbles. If
ultraviolet light is used to accomplish polymerization,
the composition must be UV transparent. This means that
the composition must be free of coloring pigments, or
20 other ultraviolet light absorbing fillers. Also,
because the process requires the compositions to
comprise a photoinitiator, the compositions tend to
yellow over time.

U.S. Patent No. 4,612,242 to Vesley et al., also
25 assigned to Minnesota Mining and Manufacturing Co.,
indicated that the white color of the Levens tape,
caused by the absence of pigment, made the tape
undesirably visible in certain applications, but that
the addition of carbon black in an amount sufficient to
30 produce a desirable black appearance would block the
ultraviolet radiation from polymerizing the mixture to a
pressure-sensitive adhesive state. Vesley et al.'s
solution to the problem was to coat the glass
microbubbles with an inorganic film, e.g. silver, having
35 a thickness that does not unduly inhibit polymerization.

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1 While the method of Vesley et al. does impart some
color to the tapes, it has certain drawbacks. It still
takes a long time to effect polymerization, making the
tapes expensive to produce. Moreover, glass
5 microbubbles having an inorganic coating are expensive,
adding to the cost of producing the tapes. Only a
limited number of colors are available in this process.

Summary of the Invention

10 The present invention provides a pressure sensitive
adhesive (PSA) tape comprising at least one carrier
layer having a composition comprising a cross-linked
polymer matrix, preferably a cross-linked PSA polymer
matrix, and more preferably, an electron beam-cured PSA
15 polymer matrix. The carrier layer further comprises low
density microspheres and at least one pigment. The
carrier layer is preferably coated on each side with a
skin layer having an adhesive polymer matrix free of
rigid low density microspheres.

20 The polymer matrix of the carrier layer is
preferably an acrylic based PSA polymer matrix or a
rubber based PSA polymer matrix. The polymer matrix
constitutes from 30% to about 90% by volume, preferably
from about 55% to about 90% by volume and more
25 preferably from about 70% to about 85% by volume of the
carrier layer, the balance being made up of fillers.

30 The low density microspheres of the carrier layer
are generally in the size range of from about 10 microns
to about 300 microns and may be made of ceramic,
polymeric, glass, carbon or other suitable material.
Mixtures of such low density microspheres may be used.
The low density microspheres may be solid, hollow or
porous, rigid or elastomeric, and tacky or nontacky.
The material of the low density microspheres, if
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1 desired, may be selected to cross-link with the polymer matrix during curing.

5 The low density microspheres are present in an amount of from about 5% to about 70% by volume of the carrier layer and preferably in an amount of from about 5% to about 45% by volume and more preferably in an amount of from about 10% to about 20% by volume of the carrier layer.

10 The pigment is present in an amount sufficient to impart the desired color to the tape. Pigment may be a solid inorganic filler such as carbon black, titanium dioxide or the like, or may be an organic dye.

15 Preferably, the carrier layer comprises fumed silica in an amount of up to about 5% by weight and more preferably in an amount of from about 1% to about 2% by weight.

20 The thickness of the carrier layer is not critical but is preferably in the range of from about 0.25 mm to about 4.0 mm and more preferably in the range of from about 0.25 mm to about 2.0 mm. The coating thickness of the rigid low density microsphere-free skin layers is preferably about 25 to about 125 grams/square meter.

25 The foam-like tapes of the present invention exhibit high conformability which arises from the low elastic memory of the carrier layer. The tapes also exhibit high failure strain, high cleavage peels and tensile adhesion, and good gasoline and moisture resistance. If inherently tacky PSA low density microspheres are used, the tapes also exhibit greatly improved cold temperature properties.

30 The invention further provides a process for rapidly producing curable bubble-free PSA tapes as described above in virtually any practical thickness. The adhesive sheet materials are produced from an adhesive composition containing from about 40% to about

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1 80% solids, i.e., about 20% to about 60% by volume
solvent. The "solids" portion of the adhesive
composition comprises a curable adhesive polymer matrix
and may comprise one or more fillers such as pigments,
5 solid, hollow or porous low density microspheres and the
like. The curable adhesive polymer matrix comprises one
or more monomers which have been at least partially
polymerized and preferably completely polymerized.

10 In the process, the adhesive composition is
introduced into a twin screw extruder through an
upstream feeding unit at the entrance of the extruder
barrel. The rotating screws of the extruder convey the
adhesive composition downstream through the extruder
15 barrel from the feeding unit to a die at the downstream
end of the extruder. At the downstream end, the
adhesive composition exits the extruder through the die.

At one or more locations downstream of the feeding
unit, the extruder comprises a solvent removal unit.
The solvent removal unit comprises a barrel section
20 having a vent opening. A conduit or duct encloses the
vent opening and extends from the vent opening to a
vacuum pump. The vacuum pump is arranged to reduce the
atmospheric pressure within the duct, the vent opening
and the barrel section to thereby draw off solvent
25 present in the adhesive composition moving through that
barrel section.

In the process, the temperature of the material
passing through the barrel section of the solvent
removal unit and the atmospheric pressure within the
30 barrel section are adjusted to cause the solvent in the
material to evaporate and be drawn off without drawing
any of the adhesive composition through the vent
opening. Elevated temperatures of from about 100°C to
about 160°C in combination with an atmospheric pressure
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1 of from about 50 to about 100 torr are presently preferred.

5 In a preferred embodiment of the invention, the extruder is provided with two or more solvent removal units. Each solvent removal unit comprises a barrel section having a vent opening which is connected by a duct to a vacuum pump to reduce the atmospheric pressure within the barrel section. In an embodiment involving
10 three solvent removal units, it is presently preferred that approximately 80% of the solvent in the composition is removed as the adhesive composition passes through the barrel section of the first solvent removal unit; an additional 18% to 19% of the solvent is removed as the adhesive composition passes through the barrel section
15 of the second solvent removal unit; and another 1% to 2% is removed as the adhesive composition passes through the barrel section of the third solvent removal section.

20 After the solvent is removed, the adhesive composition preferably exits the die of the extruder onto a backing film or the like.

In a particularly preferred embodiment of the invention, there is provided a co-extrusion process for making a laminated PSA composition having at least one first layer, e.g. a carrier layer, of a particular first
25 polymeric composition which may or may not be a PSA composition and at least one second layer, e.g. a skin layer, of second composition which is a PSA composition. The process utilizes two twin screw extruders, each having one or more solvent removal units as described
30 above. A first mixture comprising the first composition and solvent is introduced into the first extruder. Simultaneously, a second mixture comprising the second composition and solvent is introduced into the second extruder. In each extruder, the solvent is stripped by
35 the solvent removal unit, and the composition is passed

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1 into a single sheet die from which the laminated PSA
sheet material is extruded.

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1 Brief Description of the Drawings

These and other features and advantages of the present invention will be better understood by reference to the following detailed description when considered in
5 conjunction with the accompanying drawings wherein:

FIG. 1 is a longitudinal, cross-sectional view of an extruder useful in the practice of the present invention;

10 FIG. 2 is a transverse cross-sectional view of the extruder taken through lines 2-2;

FIG. 3 is a transverse cross-sectional view of the barrel showing an alternatively preferred feed unit;

FIG. 4 is a schematic view of the extruder barrel showing the screw profile; and

15 FIG. 5 is a fragmentary cross-sectional view showing an adaptor for extending a laminated PSA sheet.

FIG. 6 is a fragmentary cross-sectional view of a preferred die assembly.

20 FIG. 7 is a top cutaway view of the die assembly of FIG. 6 taken along lines 7-7.

FIG. 8 is a front view of the die assembly of FIG. 6.

FIG. 9 is a schematic view of an extruder barrel showing a particular screw profile; and

25 FIG. 10 is a schematic view of an extruder barrel showing another particular screw profile.

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1 Detailed Description of the Invention

5 In a particularly preferred embodiment of the present invention, there is provided a foam-like double-coated PSA tape having excellent conformability, high failure strain, high cleavage peels and tensile adhesion, and good solvent resistance. The double-coated PSA tape is a composite structure comprising a middle carrier layer and a skin layer on each side of the carrier layer.

10 The thickness of the carrier layer is not critical but is preferably from about 0.25 mm to about 4.0 mm and more preferably from about 0.25 mm to about 2.0 mm. Likewise the coating thickness of the skin layer is not critical but is preferably in the range of from about 25 g/m² (approximately 1 mil) to about 125 g/m² (approximately 5 mils).

15 Carrier layers having a thickness greater than about 4.0 mm are not preferred because they contain excess material which is generally not needed for most applications. Further, thicker carrier layers tend to be more visible in a particular application and are therefore not generally preferred because they are less aesthetically pleasing. Such carrier layers also require higher voltages for electron-beam curing.

20 Carrier layers having a thickness less than about 0.25 mm are not preferred because they tend to exhibit insufficient conformability and thus are less suitable for use with irregular surfaces. Thicknesses in the range of about 0.25 mm to about 2.0 mm are suitable for most applications.

25 The skin layers typically exhibit better adhesion than the carrier layer and thus enhance the overall adhesion of the tape. Skin layers having a coating thickness less than about 25 g/m² are not preferred because no significant benefit is seen. Skin layers

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1 having a coating thickness greater than about 125 g/m²
are not preferred because no additional benefit is seen
with greater thickness.

5 In accordance with the invention, the carrier layer
comprises a cross-linked polymer matrix, low density
microspheres, and at least one colored pigment. The
polymer matrix is preferably an acrylic based PSA
polymer matrix or a rubber based PSA polymer matrix.
Cross-linking is preferably accomplished by electron-
10 beam curing.

Acrylic-based PSA polymer matrices generally
comprises one or more first monomers, which if
homopolymerized, would have a glass transition
temperature of less than about -25°C based on the total
15 weight of the monomers. Examples of such monomers
include alkyl acrylates such as butylacrylate,
propylacrylate, 2-ethyl hexylacrylate, isooctyl
acrylate, isodecylacrylate, and the like. The balance
of the monomer system may be comprised of second
20 monomers which, if homopolymerized, would have a glass
transition temperature greater than -25°C, normally
greater than about 10°C. Among such monomers there may
be mentioned ethyl acrylate, alkyl methacrylate such
as methyl methacrylate, ethyl methacrylate, butyl
25 methacrylate and the like; co-polymerizable vinyl-
unsaturated monomers such as vinyl acetate, vinyl
propionate and the like; and styrenic monomers such as
styrene, methyl styrene and the like, unsaturated
carboxylic acids such as acrylic acid, methacrylic acid,
30 itaconic acid, fumaric acid, and the like; acrylamide,
vinyl caprolactam and the like. Suitable polymers are
described, for example in co-pending U.S. Patent
Application Serial Number 916,717, which is incorporated
herein by reference.

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1 Rubber-based PSA polymer matrices useful in the
practice of the present invention may be formulated as
solvent, hot melt, or emulsion, with holt melt or
5 solvent based adhesives presently being preferred. The
PSA matrices employed are normally based on di-block and
tri-block polymers and mixtures thereof. Other resin-
modified elastomers could be used. The matrix polymer,
to be functional, should have a net effective glass
10 transition temperature of from about 15°C to about 70°C
below the use temperature, preferably from about 35°C to
about 70°C below the use temperature. Rubber based
adhesive suitable for use in the present invention are
described in U.S. Patent Nos. 3,239,478 to Harlan,
15 4,152,231 to St. Clair, et al., 3,676,202 to Korpman,
3,783,072 to Korpman, 3,932,328 to Korpman and 4,028,292
to Korpman and partially in U.S. Patent application
Serial No. 896,127, all of which are incorporated herein
by reference.

20 It is preferred that the polymer matrix of the
carrier layer be a PSA polymer matrix. It is
understood, however, that other materials may be used as
the polymer matrix. Non-PSA polymers suitable for use
as the polymer matrix of the carrier layer include
polyethylene, ethylene propylene rubbers, neoprene,
25 butyl rubber and the like.

Cross-linking of the polymer matrix is preferably
accomplished by electron-beam curing. Accordingly, it
is understood that other electron beam curable polymer
materials such as electron beam curable silicones, may
30 also be used, if desired.

While not presently preferred, other suitable
methods for curing the polymer matrix may be used. For
example, if the polymer matrix comprises a heat-
activatable cross-linking agent, curing by the
35 application of heat may be used. If the polymer matrix

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1 also contains a microwave absorbing agent, microwave
radiation may be used to effect curing. Because of the
presence of pigment in the polymer matrix, ultra-violet
radiation is not an appropriate method for curing the
5 polymer matrix of the present invention.

The polymer matrix is present in the carrier layer
in an amount of from about 30% and preferably from about
55% to about 90% by volume, and more preferably in an
amount of from about 70% to about 85% by volume. Stated
10 in another way, the total amount of fillers should be at
least 10% by volume and no more than about 70%, and
preferably no more than about 45% by volume, and more
preferably in the range of from about 15% to about 30%
by volume. Carrier layers having more than about 45
15 volume percent fillers, or more than about 70 volume
percent fillers, if very low density fillers, e.g.,
penolic hollow microspheres, are used, tend to exhibit
low elongation and high modulus and not generally
suitable for PSA applications. Likewise, carrier
20 layers having less than about 10% by volume fillers are
not preferred because the resultant tapes generally
possess too low of a modulus.

Carrier layers having from about 15 to about 30
volume percent fillers are most preferred because such
25 compositions tend to exhibit the best combination of
properties such as elongation and tensile strength.

The carrier layer also comprises from about 5% to
about 70% by volume, preferably 5% to about 45% by
volume, and more preferably from about 10% to about 20%
30 by volume low density microspheres. The low density
microspheres tend to reduce the density of the carrier
layers, generally improve peel adhesion and thereby
improve conformability and also improves the strength
properties, i.e. the combination of elongation and
35 tensile strength of the layer.

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1 The low density microspheres may be solid, hollow
or porous and rigid or elastomeric. The low density
microspheres may be made of any suitable material
including glass, ceramic, polymeric and carbon
5 materials.

 Polymeric low density microspheres may be made of
rigid materials or elastomeric materials. Suitable
rigid polymeric materials include thermosetting
polymers, e.g., phenolic polymers, or thermoplastic
10 polymers, e.g., polyvinylidene chloride acrylonitrile
copolymers (PVDC copolymers). It is expected that
thermoplastic polymer microspheres will cross-link and
graft to the polymer matrix when electron-beam
radiation is used to cure the polymer matrix. By cross-
15 linking the low density microspheres and grafting to the
polymer matrix, properties such as tensile strength
could be improved.

 Preferred elastomeric low density microspheres are
made of a PSA material and exhibit a very low glass
20 transition temperature (T_g), are infusible, insoluble
and inherently tacky. Such elastomeric low density
microspheres can be made, for example, by suspension
polymerization as disclosed in U.S. Patent Nos.
3,691,140 to Silver, 3,857,731 and 4,166,152 to Baker et
25 al., and 4,495,318 to Howard, and U.S. Patent
Application Serial No. 138,509, all of which are
incorporated herein by reference.

 Infusible low density microspheres disclosed in
U.S. Patent Nos. 4,735,837, 4,049,483, 4,645,783,
30 4,624,893, 4,636,432, 4,598,112, Japanese Patent No.
61258854, all of which are incorporated herein by
reference, are also suitable for use in the present
invention.

 It has been found that the incorporation of
35 elastomeric low density microspheres in the carrier

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1 layer improves the low temperature performance of the
foam tapes of the present invention, particularly in
cold slam tests, e.g., Fisher Body Materials Testing
(FBMT) 45-89, at temperatures of, for example, -20°C and
5 -30°C.

As used herein, "low density microspheres" include
rigid microspheres having a density of less than about
1.0 g/cc and elastomeric microspheres having a density
of less than about 1.5g/cc. Accordingly, rigid
10 microspheres made of glass, ceramic or other material
and having a density greater than about 1.0 g/cc and
elastomeric microspheres having a density greater than
about 1.5g/cc are not preferred. Such high density
microspheres tend to adversely increase the density of
15 the carrier layer requiring higher electron-beam
voltages for curing.

Hollow microspheres, which are generally available
in a wide variety of densities and crush strengths, are
presently preferred. Ceramic hollow microspheres are
20 particularly preferred because they exhibit high crush
strength and tend to be less expensive than glass,
polymeric or carbon hollow microspheres.

The size, i.e., the average diameter, of the low
density microspheres is preferably from about 10 to
25 about 300 microns. Low density hollow microspheres
having a diameter less than about 10 microns may be
suitable but are not presently commercially unavailable
for evaluation.

Low density hollow microspheres having an average
30 diameter greater than about 300 microns are not
preferred at the present time due to a lack of
commercial availability and because they are expected to
exhibit a undesirable low crush strength.

If the carrier layer comprises rigid, low density
35 microspheres made of, for example, glass or ceramic

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1 having a density of from about 0.2 to about 1.0 g/cc, it
is preferred that the loading of low density
microspheres not exceed about 45% because carrier layers
with higher loadings tend to exhibit very low
5 elongation. If low density rigid microspheres having a
density less than about 0.2g/cc, e.g., hollow phenolic
microspheres, are used, the loading may be as high as
about 70% by volume. If low density elastomeric
microspheres are used, loadings as high as about 70% by
10 volume may be used.

Carrier layers having less than about 5 volume
percent low density microspheres of any kind are not
preferred because the benefit of the low density
microspheres is insufficiently realized, e.g., the peel
15 and shear adhesion tends to be too low. Moreover, the
density of the carrier layer increases as the volume
loading of the low density microspheres decreases, and
thus, low loadings requires a higher electron beam
voltage for curing. Consequently, such carrier layers
20 tend to be more expensive to produce. Volume loadings
between about 10% to about 20% are most preferred
because carrier layers having such loadings tend to
exhibit the optimum combination of elongation and
tensile strength and other physical properties.
25 Particularly preferred carrier layers have from about
15.0% to 20.0% by volume low density microspheres.

At least one pigment is present in the composition
to give color to the tape. Solid particulate pigments
tend to improve strength characteristics, i.e. increase
30 the tensile strength and reduce the elongation of the
tape. As used herein, the term "pigment" refers to any
coloring agent compatible with or dispersible in the
polymer matrix. The pigments may be solid particles
such as carbon black and other particulate pigments or
35 titanium dioxide or organic dyes such as phthalocyanine

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1 green sold by American Hoechst or 2915 dianisidine
orange sold by Harshaw Chemical. The particular type of
pigment used will depend upon the color desired. For
example, carbon black may be used if the desired color
5 is black. Titanium dioxide may be used if the desired
color is white.

The particle size range and the loading of the
pigment depends on the type of pigment utilized. For
carbon black, a loading of up to about 5% by weight may
10 be used. Loadings above 5% by weight are not preferred
because carbon black tends to decrease the elongation at
break. Loadings as low as about 0.25% by weight are
found to be sufficient to impart a suitable black color
to the tape. With respect to carbon black, any suitable
15 commercially available carbon black may be utilized. A
particularly preferred carbon black is Monarch 700
carbon black, manufactured by Cabot Corporation.

In addition to the low density microspheres and
pigment, it is preferred that the carrier layer comprise
20 a filler such as fumed silica. Fumed silica lowers the
elongation and increases the tensile strength of the
carrier layer. Accordingly, the preferred amount of
fumed silica is selected to provide the best balance of
high elongation and high tensile strength. The fumed
25 silica could be replaced by carbon black, if desired.

The fumed silica is preferably present in an amount
of up to about 10% by weight of the carrier layer.
Loadings greater than about 10% tend to result in a
carrier layer which is too stiff and insufficiently
30 conformable for most applications. Volume loadings of
from about 3% to about 5% by weight have been found to
impart the best combination of tensile strength and
elongation and are hence presently preferred.

Small, rigid high density solid microspheres having
35 a density greater than 1.0 g/cc and a size or average

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1 diameter of less than about 10 microns and preferably
from about 0.1 to about 5 microns may be used as an
alternative to or in combination with fumed silica to
5 lower the elongation and increase the tensile strength
of the carrier layer. The small, rigid, high density
solid microspheres may be present in an amount of up to
about 5% by weight. Above about 5% by weight, the
carrier layer tends to become too stiff. It is
10 presently preferred that the small, rigid, high density
solid microspheres be present in an amount of from about
1% to about 2% by weight.

It is understood that the preferred loadings of the
various above-mentioned fillers are dependent upon the
precise characteristics which are sought and on the
15 amounts of the other fillers present in the carrier
layer. For example, a relatively high loading of solid
fillers, e.g., fumed silica or small, rigid, high
density microspheres may be preferred if the loading of
low density microspheres is low. Lower loadings may be
20 preferred if the amount of microspheres is high.

It is further understood that many other fillers,
e.g., calcium carbonate, china clay, etc., may be
incorporated into the carrier layers as desired.

25 The skin layers are preferably unfilled layers of
an adhesive polymer matrix or, less preferably, may be
an adhesive polymer matrix filled with pigment. The
skin layer contains no low density microspheres. The
polymer matrix of the skin layer may be any polymer
matrix which exhibits good adhesion with the carrier
30 layer. Preferred adhesive polymer matrices include PSA
polymer matrices and heat activatable adhesive polymer
matrices.

35 The carrier layers of the present invention may be
prepared by any suitable method. For example, a mixture
of the polymer matrix, fillers and solvent may be coated

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1 onto a backing film to a desired thickness. The solvent
is then removed by drying before curing. Alternatively,
a mixture comprising the polymer matrix and fillers and
without solvent may be extruded as such a sheet or the
5 like. A calendaring process may also be used.

In a particularly preferred process, the carrier
layer is made by first preparing an adhesive composition
containing the polymer matrix, solvent for the polymer
matrix and the desired fillers. The composition is
10 introduced into an extruder and conveyed through the
extruder by the rotating screws. While in the extruder,
the solvent is removed by vacuum evaporation in one or
more solvent removal units. An essentially solvent-free
composition is then extruded from the extruder. As used
15 herein, "solvent-free" means a composition having less
than about 2% by volume solvent.

Exemplary solvents include ethyl acetate,
isopropanol, ethanol, hexane, heptane and toluene. The
purpose of the solvent is to reduce the viscosity of the
composition so that it may be easily handled in bulk,
20 e.g., readily poured from one container to another. An
amount of solvent sufficient to reduce the viscosity to
less than about 100 pascal-seconds is presently
preferred. For most compositions, an amount of solvent
that provides a solids content of from about 40% to
25 about 80% is sufficient for this purpose. That is,
compositions having more than about 80% solids are not
preferred because the viscosity remains undesirably
high. Compositions having less than about 40% solids
30 are not preferred because they contain excess solvent,
i.e. more than enough solvent to reduce the viscosity to
an easily workable level, and the excess solvent must be
removed in the process. The particular viscosity
desired will depend on the method by which the
35 composition is introduced into the extruder.

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1 The particular amount of solvent required to
achieve a desired viscosity will depend on the temper-
ature of the composition. Accordingly, the composition
is preferably heated to minimize the amount of solvent
5 required to achieve the desired viscosity which, in
turn, minimizes the amount of solvent that has to be
removed in the process. Temperatures slightly below the
boiling point of the composition are preferred.

10 With reference to FIG. 1, there is shown
schematically an apparatus suitable for use in
practicing the present invention. The apparatus
comprises a twin screw extruder 10 with three solvent
removal units 11, 12, and 13 for removing solvent from
an adhesive composition traveling through the extruder
15 10. A sheet die 14 is mounted at the downstream end of
the extruder 10. In such an arrangement, a solvent-
free adhesive composition may be extruded in the form of
a sheet.

20 In the embodiment shown, a backing film or web
feeding unit 16 is provided for applying a release film
to one side of the extruded sheet. A conveying unit 17
is also shown for carrying the extruded sheet material
away from the extruder 10. It is understood that the
processing of the extruded sheet, e.g., application of a
25 backing film conveying away from the extruder,
subsequent curing, etc., may be accomplished by any
suitable conventional method. Subsequent curing by
electron beam radiation is currently preferred.

30 The twin screw extruder 10 may be any suitable
commercially available twin screw extruder which is
modified to include one or more solvent removal units.
For example, extruders manufactured by Berstorff
Corporation of West Germany, have been found to be
suitable for use in the practice of this invention.
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1 With reference to FIG. 2, the extruder 10 comprises
a housing or barrel 18 having a pair of side-by-side
generally parallel and cylindrical overlapping bores 19
forming a barrel chamber 22, in which a pair of co-
5 rotating intermeshing screws 21 are mounted. While it
is presently preferred that the extruder 10 have co-
rotating screws 21, it is understood that extruders
having counter-rotating screws may also be used. It is
also understood that arrangements in which the screws do
10 not intermesh can also be used. For compositions
involving breakable low density microspheres, e.g,
hollow glass microspheres, use of tangential screws may
reduce breakage of the microspheres.

15 The barrel 18, preferably comprises multiple sec-
tions. The combination and arrangement of barrel
sections are selected to accomplish specific tasks. The
barrel sections may completely enclose the screws or
have openings for feeding, venting and the like. Each
section of the barrel is provided with a heating means
20 so that the material within that barrel section may be
heated to a desired temperature.

25 Likewise, the screws 21 preferably comprise
multiple elements designed to accomplish the particular
tasks such as mixing, conveying, building pressure and
the like. The combination and arrangement of screw
elements are selected to accomplish desired tasks in a
particular order.

30 In the embodiment shown, the barrel 18 comprises
seven sections. The first section 24 is part of a
feeding unit 26 for introducing material into the
extruder. The feeding unit 26 comprises a large feed
hopper 27 which empties directly into the chamber 22 of
the first barrel section 24 through an entrance port 28,
as shown in FIG. 3. While not shown in the drawing, a
35 feeding or metering unit may be provided at the entrance

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1 port 28 to control the rate of flow of material from the
hopper 27 into the barrel 18.

5 It is understood that, rather than mixing the
fillers, polymer matrix and solvent together and then
introducing the mixture into the extruder, one or more
of the fillers can be introduced separately. If added
separately, it is preferred that the filler be added to
composition already in the barrel. This provides mixing
10 between the polymer matrix and the filler and reduces
clumping and possible crushing of the filler by unwetted
screws. As an example, the composition comprising
polymer matrix and solvent may be introduced in a first
feed unit at the first barrel section and the solid
15 fillers may be introduced in a second downstream feed
unit.

Alternatively, the filler and the composition
comprising polymer matrix and solvent may be added in
the same barrel section in an arrangement as shown in
FIG. 3. In such an arrangement, the composition
20 comprising polymer matrix and solvent is introduced into
the barrel chamber 22 through an entrance port 29 at the
bottom of the barrel 18. The composition thus
introduced tends to puddle at the saddle area 30 of the
barrel 18 wetting the screws 21 as they rotate. The
25 filler is introduced into the extruder at the top of the
barrel 18, for example through the feed hopper 27,
directly onto the wetted screws.

If the separately added solid filler comprises
breakable low density microspheres, e.g., hollow glass
30 microspheres, it is presently preferred to add the
filler at the downstream end of the extruder, i.e., at a
location downstream from the solvent removal units, to
reduce breakage of the low density microspheres. In
such an embodiment, the screws, if desired, may be
35 intermeshing through the solvent removal units to

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1 enhance devolatilization of the adhesive composition and
then become tangential at the downstream and of the
extruder where the breakable low density microspheres
are added.

5 The first, second, and third solvent removal units
11, 12, and 13 are located downstream of the feeding
unit at the fourth, fifth and sixth barrel sections 31,
32, and 33 respectively. As shown in FIG. 2, each of
the fourth, fifth and sixth barrel sections 31, 32 and
10 33 has a large vent opening 34 at the top of that barrel
section. A duct 36 extends from the vent opening 34 to
a vacuum pump 37 for reducing the atmospheric pressure
within the duct 36, vent opening 34 and that barrel
section. In the embodiment shown, each solvent removal
15 unit has a separate vacuum pump. It is understood that
two or even three ducts may be joined so that only one
or two vacuum pumps are required to reduce the
atmospheric pressure in all three solvent removal units.
Solvent removed is preferably collected, for example by
20 condenser 38.

With reference to FIG. 4, there is shown a
preferred screw profile suitable for use in the present
invention. In the first barrel section 24, the screws
21 have a return scroll element 40 which prevents
25 material from back flowing into the drive unit 39 (FIG.
1). The portions of the screws 21 extending through the
remainder of the first barrel section 24, where the
material is introduced into the extruder 10, and the
second barrel section 25, comprise open chamber
30 conveying elements 41 which rapidly transport the
material downstream. Conveying elements 41 have very
thin flights and therefore tend not to generate a
significant amount of back pressure.

In the third barrel section 29, the screws 21 are
35 designed to build pressure. In the embodiment shown,

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1 this is accomplished with a series of mixing elements 42
followed by closed chamber conveying elements 43 having
large, thick flights. The conveying elements 43 are
followed by another series of mixing elements 42 and
5 then a blister 44. The blister 44 has a large diameter
to restrict the flow of material past it.

Material which has squeezed past the blister 44 is
conveyed rapidly through the fourth barrel section 31
and past vent opening 34 by a series of open chamber
10 conveying elements 41. Such an arrangement maximizes
the surface area of the material traveling through the
fourth barrel section 31 and hence maximizes the removal
of solvent by the first solvent removal unit 11.

At about the beginning of the fifth barrel section
15 32, before the next vent opening 34, the screws 21
comprise another series of mixing elements 42 followed
by another blister 44.

In the present process, the blisters 44 may all be
the same size, there being less of a need to increase
20 the size of downstream blisters because the material
becomes more viscous as it travels through the extruder.
As material becomes more viscous it tends to build
pressure in the extruder more readily. This tends to be
the opposite of most extruding processes in which solid
25 materials are fed into the extruder and the viscosity of
the material decreases as it becomes hotter.

Material squeezing past the blister 44 is again
carried rapidly past the vent opening 34 by open chamber
conveying elements 41. Again, the arrangement maximizes
30 the surface area of the material exposed to the
atmosphere in the fifth barrel section 32 and hence
maximizes solvent removal.

A similar arrangement of screw elements is provided
in the sixth barrel section 33 except that kneading
35 elements 45 are preferably used rather than mixing

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1 elements 42. The kneading elements in combination with
a blister build pressure similar to the mixing elements
and blister but also tend to remove any last traces of
5 air bubbles in the composition. In the seventh barrel
section 35, the screw 21 comprises open chamber
conveying elements 41 which convey the material to the
die.

10 The above screw profile provides an arrangement
wherein back pressure is built up before each vent
opening and then released as the material travels past
the vent opening 34 to expose as much of the composition
as possible to the atmosphere. While such an
arrangement is presently preferred, it is understood
15 that other arrangements may be used. It is also
understood that other screw elements may be used to
provide the desired pressure changes with the extruder.

20 The screw profile is preferably designed to
maximize the surface area of the composition passing
through the barrel sections of the solvent removal
units. In addition to the surface area, solvent removal
is dependent on the temperature of the composition, the
atmospheric pressure within that barrel section and the
residence time of the composition within that barrel
section which, in turn, depends on the feed rate.

25 For a given feed rate, temperature and pressure are
adjusted to maximize solvent removal without drawing any
of the composition through the vent opening. Elevated
temperatures in the range of from about 80°C to about
150°C in combination with pressures of from about 50
30 torr to about 150 torr are presently preferred.

In the embodiment shown in FIG. 1, material is
extruded as a thin sheet directly onto backing films or
webs. Rolls 47 of backing film are mounted above and
below the sheet die 14 of the extruder 10. The thin
35 sheet of material and the film from the rolls 47 pass

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1 between a pair of small rollers 48 and extend across a
conveyor 49 which carries the sheet material away from
the extruder 10 for curing.

5 One of the surprising results of the present
process is that the extruded sheets exhibits a lower-
than-expected free monomer level. In conventional
drying processes, the residual free monomer level is
about 0.5 to 2%. Moreover, the residual free monomer
10 level tends to substantially increases as the thickness
of the sheets increases. In many applications,
particularly medical applications, such free monomers
are considered undesirable impurities. In the present
process, a residual free-monomer level of 0.1% and below
can be achieved. Accordingly, products made by the
15 present invention would offer distinct advantages in
such applications.

Another surprising result is that, even at
thicknesses as great as 1mm or more, the extruded sheet
is bubble free.

20 It is apparent that the number of solvent removal
units may vary. That is, a single solvent removal unit
may be used in certain applications, particularly those
which do not require a solvent-free extrudate.
Alternatively, many solvent removal units may be used,
25 for example, if it is desired to achieve a very low
solvent or residual monomer level.

It is apparent that other compounding ingredients,
such as plasticizers, tactifying resins, fillers, cross-
linking agents and the like may be added to the extruder
30 to mix with the adhesive composition.

The present process may also be utilized in a co-
extrusion process to co-extrude thin unfilled adhesive
skin layers over both sides of the carrier layer. Such
a process utilizes two twin screw extruders. Each of
35 the extruders are set up generally as described above

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1 and comprise at least one feeding unit and at least one
solvent removal units. However, both extruders feed
material into a single die through an adaptor.

5 With reference to FIG. 5, there is shown an adaptor
50 suitable for such an application. The adaptor 50
comprises a first pipe 51 extending forwardly from the
first extruder 52 to the back of a sheet die 53 for
10 carrying material from the first extruder 52 to the die
53. A larger diameter second pipe 54, having a closed
rearward end 55 is mounted concentrically around the
first pipe 51 adjacent to the die 53. The diameter of
the second pipe 54 is selected to form an annular space
15 56 around the first pipe 51. A third pipe 57 connects
the second extruder 58 to the second pipe 54 and carries
material from the second extruder 58 to the annular
space 56. Both of the first and second pipes 51 and 54
open into the interior of the die 53.

In the process, a first composition comprising
first polymer matrix, solvent and fillers, as required
20 for forming the carrier layer, is introduced into the
first extruder 52. Simultaneously, a second adhesive
composition comprising second polymer matrix, e.g., a
PSA polymer matrix, and solvent is introduced into the
second extruder 58. Solvent is removed from each
25 composition by the solvent removal units of the
extruders as described above. A solvent-free first
composition from the first extruder 52 flows into the
die through the first pipe 51. Likewise, solvent-free
second adhesive composition from the second extruder 58
30 flows through the third pipe 57 and then the second
pipes 54 and into the die as a concentric ring around
the first adhesive composition. In the die, the
adhesive compositions are flattened out and extruded in
a laminated sheet construction, the first adhesive
35 composition forming a middle carrier layer and the

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1 second adhesive compositions forming the top and bottom
skin layers.

5 The skin layer increases the tack or initial
adhesion of the tape. As an alternative to co-extruding
a skin layer covering the entire carrier layer, the skin
layer may be "co-extruded" with the carrier layer as
strips or patches at the surface of the carrier layer.

10 With reference to FIGS. 6, 7 and 8, there is shown
a preferred die assembly for co-extruding one adhesive
material with strips of a second adhesive material on
its surface. The die assembly comprises a conventional
sheet die 61. The top and bottom plates 62 and 63 of
the die 61 comprise a series of cylindrical bores 64. A
small opening 66 connects each bore 64 with the interior
15 of die 61.

The material of the carrier layer is processed
through a first extruder 67 having solvent removal units
as described above and is introduced into the back of
die 61. The second material, e.g., a PSA polymer matrix
20 free of microspheres and filler, is introduced into the
die through an adaptor 68. The adaptor 68 comprises a
pipe 69 and upper and lower manifolds 71 and 72. The
upper and lower manifolds 71 and 72 cover the bores 64
in the top and bottom plates 62 and 63 of the die 61.

25 The second material is introduced into the pipe 69,
flows into the upper and lower manifolds 71 and 72, into
the bores 64 and through the openings 66 into the
interior of the die. In such an arrangement the second
adhesive is "co-extruded" with the carrier material as
30 strips 73 on the surface of the extruded carrier
material 74. The width of the strips 73 depends on the
diameter of the openings 66. Likewise the depth of the
strips depends on the rate at which the second material
is introduced into the die through the openings. If
35 desired, the second material may be pulsed into the die,

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1 thus forming broken strips or patches of the second
material at the surface of the carrier material. It is
apparent that the outer surface of the strip 73 is
generally coplanar with the exposed surface of the
5 carrier material 74.

 This embodiment offers more latitude in selecting
the second material than a co-extrusion process in which
the second material forms a skin layer covering the
entire surface of the carrier layer. With a skin layer
10 covering the entire surface, the materials of the skin
and carrier layers must exhibit good adhesion to each
other to prevent delamination. Formation of a skin
layer comprising strips makes that requirement less
critical because here is greater contact area between
15 the second material and the carrier material. Also, if
the carrier layer comprises a PSA polymer matrix, a
substantial amount of the surface of the carrier layer
is exposed and thus able to form a permanent bond with
the substrate to which it is applied.

20 The second material may be processed in a second
extruder as described above and introduced into pipe 69
from the second extruder. Alternatively, if the second
material is an acrylic or rubber based hot melt adhesive
or the like, it may be introduced into the pipe 69 by
25 means of a gear pump or the like. Hot melt adhesives
are presently preferred as they eliminate the need for a
second extruder.

 If desired, the backing film may be extruded
simultaneously with the adhesive sheet and applied
30 directly to the surface of the adhesive sheet. Also,
rather than a co-extrusion process as described above,
it is apparent that the tape and backing film may be co-
extruded using a conventional blow film extrusion
process.

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1 For certain substrates such as PVC side moldings,
the foam-like tape may be co-extruded with the substrate
in a single operation.

5 The foam-like tape sheet or tape thus produced is
preferably cured, i.e. cross-linked, by electron beam
radiation. The carrier layer may be cured prior to or
after lamination of the skin layers. Typical electron
beam radiation levels range from about 10 to about 100
kiloGray (kGy) and are preferably from about 30 to about
10 60 kGy.

The foam-like tapes of the present invention
exhibit an excellent combination of rheological,
adhesion and performance properties. For acrylic based
PSA tapes, tensile strength, as measured by ASTM D1708,
15 is typically in the range 0.5 to 1.3 megapascal.
Elongation, as measured by ASTM D1708, is preferably
from about 500 to about 1500 percent or greater. For
rubber based PSA tapes, the tensile strength is
typically from about 0.7 to about 2.0 megapascals and
20 the elongation is from about 500 to about 2500%.

For the acrylic based PSA tapes of the present
invention, the storage modulus (G'), measured at 0.01
radians frequency at 25°C is at least 10^4 pascals and
preferably at least 4×10^4 pascals after electron beam
curing. The loss modulus (G'') generally measured at
25 0.01 radians frequency at 25°C is at least 10^4 pascals
and preferably at least 4×10^4 after electron beam
curing. When measured at 100 radians frequency, G' and
 G'' are both less than about 2×10^6 pascals.

30 The peel adhesion is preferably from about 1300 to
about 3000 Newtons/m or greater for acrylic based PSA
tapes and from about 3000 to about 12000 Newtons/meter
or greater for rubber based PSA tapes measured by PSTC
No. 3. In the test one side of the tape is laminated to
35 soft 0.05 mm aluminum foil and then tested after

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1 laminating the other side to the substrate with a 6.8 kg
roller, two passes, and then waiting for twenty minutes.
If the peel adhesion is greater than 350° Newtons/meter,
5 a 5 mil polyester film is used rather than aluminum
foil.

The shear adhesion as measured by Fisher Body
Materials Specification (FBMS) Test Method (TM) 45-124,
is preferably at least 500 grams for acrylic PSA tapes.

10 In the following examples, certain designations and
trade names are used. Adhesive A is an electron beam
curable acrylic solution adhesive comprising butyl
acrylate, 2-ethyl hexyl acrylate and acrylic acid in a
45:41:19 mole ratio. Adhesive B is an electron beam
15 curable acrylic solution adhesive comprising butyl
acrylate, 2-ethyl hexyl acrylate and acrylic acid. RB
designates a rubber based solution adhesive comprising
about 19.3% by weight styrene-butadiene-styrene linear
copolymer containing about 31% styrene, about 16.1% by
weight styrene-butadiene copolymer, about 25.8% by
20 weight alpha pinene tackifier, about 32.3% by weight
rosin ester tackifier, and about 6.4% by weight of a
compatible aromatic liquid resin. A-16-500 designates
hollow glass microspheres marketed by Minnesota Mining
and Manufacturing Co. having a true particle density of
25 about 0.2 g/cc and a size of 20-130 microns. Q-cel 500
designates hollow glass microspheres marketed by P.Q.
Corp. having a true particle density of about 0.2 g/cc
and a size of 10 to 115 microns. Cab-O-Sil M5
designates fumed silica from Cabot Corp. Monarch 700
30 carbon black is sold by Cabot Corp. BJO 0930 is a trade
designation of Union Carbide Corp. for hollow phenolic
microspheres having an average particle size of 40
microns. SF-14 is a trade designation of PA Industries
for hollow ceramic microspheres having a density of
35 0.7g/cc and a particle size of 10-100 microns.

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Example I

The following compositions listed in Table I were prepared by adding the fillers to a solution of the polymer in ethylacetate/isopropanol at 50% by weight solids:

5

Table I

	<u>Composition 1</u>	<u>Composition 2</u>	<u>Composition 3</u>	<u>Com. 4</u>	<u>Com.5</u>
10	Adhesive A 100g (dry wt.)	100g	_____	100	100
	Adhesive B _____ (dry wt.)	_____	100g	-	-
15	Glass Hollow Microspheres A-16-500 6g	6g	6g	-	-
	Q-Cel-500 _____	---	---	-	4g
	Cab-0-Sil M5 4g	3g	4g	4g	4g
20	Carbon Black Monarch 700 _____	1g	_____	-	0.2g
	Phenolic Hollow Microsphere BJO 0930 _____	---	_____	-	2g
25	Ceramic Hollow Microsphere SF14 _____	---	_____	16g	-

30

Each of the above compositions were coated onto a release film and dried at 70°C in an oven with forced air circulation for 20 minutes and then in a vacuum oven at 70°C for one hour. A 0.8mm thick, 20cm x 20cm carrier layer was prepared by compression molding at about 110°C using a stainless steel mold. A teflon FEP film was used to prevent the adhesive from sticking to the mold. The carrier layer was then electron beam irradiated both sides at 50 kGy using a 300 KeV ESI

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1 electron beam equipment. A high performance acrylic transfer tape AS 838X manufactured by Avery was then laminated on both sides of the carrier layer.

Composition 1 involved two different samples.

5 Sample 1 was cured open face and sample 2 was cured through a FEP release film, composition 4 also involved two different samples. The skin layers of sample 2 had a coat weight of 125 G/m² rather than 50 g/m², as in Sample 1.

10 The dynamic mechanical properties of the tapes were evaluated using a Rheometrics dynamic mechanical spectrometer at 24°C at a frequency range of 0.1 - 300 rad/sec.

15 PVC side molding test bars from Standard products were wiped clean with 1:1 dilution of isopropanol and distilled water and dried at room temperature. The moldings were then primed with Tite-R-Bond 2287 from Norton Chemical and dried at room temperature for 12 hours. One side of the double coated tape was laminated onto the smooth surface of the side molding using 6.8 Kg weighted roller. Painted panels 51mm x 127mm (Inmont base coat/clear coat) were cleaned using isopropanol/ distilled water as above and dried. The release film was removed and the molding was attached to the panel using a 6.8 Kg weighted roller (two passes) with 25.4mm of the molding overhanging the edge of the panel. All panels were aged at room temperature for 72 hours prior to an exposure or testing. Cleavage peels were determined similar to Fisher Body TM 45-88. Accelerated aging involved 2 weeks at 82°C in an air oven. Humidity resistance was tested after exposing the panels with the side molding at 38°C and 95% humidity for one week. Gasoline resistance was tested by immersing the samples into gasoline for 10 seconds and 20 seconds dry off time. This was repeated three times. Cleavage peel

20
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1 was determined immediately after the final immersion.
Initial values indicate cleavage peels immediately after
the specified exposure and Final values refer to
cleavage peels after 24 hours conditioning at room
5 temperature. Creep test involve laminating a 12.7mm x
63.5mm side molding using a 6.8Kg weighted roller on a
panel as in cleavage peel test with 12.7mm overhanging
and attaching a 500g weight at the free end of the
molding and immediately placing the sample in the oven
10 for 96 hours at 70°C. Tensile strength and elongation
at break were determined using a dumbell specimen
similar to the ASTM D 1708.

The results are shown in Table II below. The
results shown are an average of at least two duplicate
15 tests.

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TABLE VI

	Composition 1 Sample 1	Comp. 2	Comp. 3	Comp. 4 Sample 1	Comp. 5 Sample 2
CLAVAGE PEEL BREAKAWAY	121 \pm 14	82 \pm 14	80 \pm 14	86	74
CONTINUOUS	36 \pm 7	32 \pm 9	30 \pm 9	28	40
ACCELERATED AGING INITIAL (N/12.7MM) BREAKAWAY	68	50 \pm 14	62 \pm 14	92	76
CONTINUOUS	25 \pm 7	40 \pm 9	30 \pm 0	62	47
FINAL (N/12.7MM) *	188 \pm 14	150 \pm 14	132 \pm 14	147	183
CONTINUOUS	62 \pm 7	60 \pm 9	60 \pm 9	89	84
HUMIDITY INITIAL (N/12.7MM) BREAKAWAY	130 \pm 14	90 \pm 14	100 \pm 14	84	70
CONTINUOUS	63 \pm 7	50 \pm 9	40 \pm 9	40	40
FINAL (N/12.5MM) 8 *	158 \pm 14	95 \pm 14	95 \pm 14	90	90
CONTINUOUS	70 \pm 7	50 \pm 9	42 \pm 9	48	50
CASOLINE INITIAL (N/12.5MM) BREAKAWAY	130 \pm 14	88 \pm 14	90 \pm 14	74	77
CONTINUOUS	38 \pm 7	30 \pm 9	30 \pm 9	36	38
CREEP (HRS) 70 DEG.C INITIAL (KG/MIL) TENSILE STRENGTH (KPa) ELONGATION (%)	166+ 39 900 700	166+ 39 900 700	166+ 39 900 700	95+ 45 - -	95+ 45 - -
STORAGE MODULUS (G') (Pa) at 0.1 radians freq. at 100 radians freq.	1.5x10 ⁵ 9x10 ⁵	- -	- -	- -	- -
LOSS MODULUS (G'') (Pa) at 0.1 radians freq. at 100 radians freq.	4x10 ⁴ 8x10 ⁵	- -	- -	- -	- -

*Final values refer to the adhesion after recovering at 23°C for 24 hours.

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Example II

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10

A rubber based carrier layer was prepared by mixing as a solution in toluene at 50% by weight solids 90.4% by weight (dry weight) RB rubber based adhesive, 0.3% by weight trimethylolpropanetrithioglycolate (TMPTG), as a cross-linking additive and 9.3% by weight (32% by volume) hollow glass A-16-500 microspheres. The solvent was stripped off by vacuum and a 0.8 mm carrier layer was prepared by compression molding. A release liner, was used to prevent the carrier layer from sticking to the sides of the mold.

15

The rubber carrier layer thus prepared was electron beam irradiated on both sides at 300 kv, open faced under nitrogen. The process was repeated and a high performance rubber adhesive transfer tape was laminated on each side of the carrier layer to provide an adhesive coat weight of about 50 g/m² and electron beam irradiated at 50 kGy dose.

20

Tensile test specimens were cut out from the uncoated and double-coated samples and tested in a manner similar to the acrylic tapes of Example I. The results are shown in Tables III and IV below.

Table III

25

<u>PROPERTIES OF UNCOATED RUBBER-BASED CARRIER</u>				
<u>LAYER</u>	<u>TENSILE</u>	<u>ELONGATION</u>	<u>180° PEEL ON</u>	
<u>EB DOSE</u>	<u>STRENGTH</u>		<u>SS</u>	<u>PE</u>
<u>(kGy)</u>	<u>(kPa)</u>	<u>(%)</u>		
0	1000	1700	2400 P	1200 P
30	1600	1600	1700 P	1100 P
50	1300	1200	1900 P	- -

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Table IV

PROPERTIES OF DOUBLE-COATED RUBBER-BASED TAPE

EB DOSE (kGy)	TENSILE STRENGTH (kPa)	ELONGATION (%)	180° PEEL ON	
			SS	PE
0	1300	1800	>3900*	2000 P
30	1700	1900	>3900*	2300 P
50	1500	1700	>3900*	2200 P

P = CLEAN PANEL FAILURE

SS = STAINLESS STEEL

PE = POLYETHYLENE

* value at which the face stock aluminum tore

Example III

A rubber-based composition was prepared as in Example II comprising 90.8% by weight (dry weight) RB adhesive, 3.6% by weight Q-cel 500 glass hollow microsphere, 1.8% by weight BJO 0930 hollow phenolic microspheres, 3.6% by weight Cab-O-Sil and 0.2% by weight Carbon Block, Monarch 700. The solvent was stripped off by vacuum and a 0.8 mm carrier layer was prepared by compression molding. A release liner, was used to prevent the carrier layer from sticking to the sides of the mold. The process was repeated and a high performance rubber adhesive transfer tape was laminated on each side of the carrier layer to provide an adhesive coat weight of about 50 g/m². Peel adhesive was measured after laminating one side of the tape to 0.127 mm Mylar.

The carrier layer by itself demonstrated a peel strength of 5300 N/m on a stainless steel substrate and 2960 N/m on a polyethylene substrate. The double coated tape exhibited a peel strength of 9400 N/m on a stainless steel substrate and 3130 N/m on a polyethylene substrate.

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1 It is expected that the above samples could be
formulated with 0.6% TMPTG for electron beam curing to
improve high temperature performance without adversely
affecting the above demonstrated peel strength.

EXAMPLE IV

5 A double-coated adhesive tape as prepared by
dissolving a solution of Adhesive A in
heptane/isopropanol alcohol (90:10) to provide a
10 composition having 47.6% by weight solids. 137 grams of
the composition was blended with 105 grams (37.2% by
weight solids in heptane) of solids tacky microspheres
prepared by the droplet suspension polymerization method
disclosed in Example 2 of U.S. patent application Serial
15 No. 138,509. The mixture was coated onto a release
liner to a thickness of about 5 mils. The solvents were
then removed by drying the film in an oven at 70°C for
20 minutes and then in a vacuum oven at 80°C for 30
minutes. A one millimeter carrier sheet was prepared by
20 compression molding the dried mixture between two teflon
FEP film using a stainless steel plate in a hydraulic
press at 100°C. The formed sheet was then irradiated at
50 KG eb dose using a 2.5 MeV electron beam device. A
skin layer of adhesive was then laminated on each side
25 of the carrier layer. The skin layer was made of
Adhesive A containing 0.2% by weight
glycidylmethacrylate comonomer and was applied in a
thickness of 50 grams per square meter and was EB cured
at 50 KGy dose.

30 The physical properties of the tape were then
evaluated for side molding application and are shown in
Table V below.

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EXAMPLE V

Another double-coated foam tape was prepared by dissolving a solution of Adhesive A in heptane/isopropanol alcohol (90:10) to provide a composition comprising 47.6% by weight solids. 179 grams of the composition was blended with 80 grams (37.2% by weight solids in heptane) of solid, tacky microspheres prepared by the droplet suspension polymerization method of Example 2 of U.S. patent application Serial No. 138,509. The solvents were removed by drying the mixture as a film in an oven at 70°C for 20 minutes and then in a vacuum oven at 80°C for 30 minutes. A one millimeter thick carrier sheet was prepared by compression molding the dry mixture between two teflon FEP films using a stainless steel plate in a hydraulic press at 100°C. The sheet was then irradiated at 50 KGy does using a 2.5 MeV electron beam device. A skin layer made of Adhesive A was laminated on both sides of the tape at a thickness of 50 grams per square meter.

The properties of the tape were evaluated for side molding application and are shown in Table V below. A cold slam test as described in FBMS 45-89 was also conducted. The tape passed the cold slam test at -20°C, with all test samples remaining on the panel after 10 slams. At -30°C, three out of four samples stayed on the panel after 10 slams.

TABLE V

<u>Example</u>	90° Peel Adhesion			72 Hrs. Rm Temp		Gasoline		Aging		<u>Creep</u>
	<u>(N/M)</u>			<u>(N/12.7 mm)</u>		<u>(N/12.7 mm)</u>		<u>(N/12.7 mm)</u>		
	B	C		B	C	B	C	B	C	
A	1370 P	93 P	45 P	91 P	46 P	110 AD/F	48 AD/F	346+Ks		
B	1200 P	71 P	36 P	80 P	38 P	130 AD/F	65 AD/F	346+Ks		

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- 1 B = Breakaway
 C = Continuous
 P = Panel failure
 AD/F = Adhesive failed from foam due to poor anchorage.

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EXAMPLE VI

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A sheet die was mounted on a ZE40-A,33L/D Berstorff extruder generally as shown in FIG. 1 having seven barrel sections. A feed unit was mounted in the first section. The screw profile of the extruder is shown specifically in FIG. 9. Here screw element 61 is a return scroll. Screw elements 62 are three-flight conveying elements, specifically Berstorff No. 60-1-3. Screw elements 63 are mixing elements, specifically Berstorff No. ZS-10-12. Screw elements 64 are two-flight conveying elements, specifically Berstorff No. 40-1-2. Screw elements 65 and 66 are blisters, specifically Berstorff 39 and 42 respectively. Screw elements 67 are kneading elements, specifically Berstorff No. KS-9-RE.

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The extruder was set up with three solvent removal units involving the fourth, fifth and sixth barrel sections respectively. Each barrel section had a large vent opening. An 1.5 to 2 inch duct connected each of the vent openings to a vacuum pump. An Ochsner vacuum pump with a suction capacity of 120 cc/hr was used in the first solvent removal unit to reduce the atmospheric pressure in the fifth barrel section and a Busch two-stage oil pump was used in the second and third solvent removal units to reduce the atmospheric pressure in the sixth and seventh barrel sections. Solvent removed by the solvent removal units were condensed and collected using a Busch condenser.

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Trial runs involving three adhesive compositions were performed. Each had 63% solids, the remainder being ethylacetate solvent. The solids of the first

-40-

1 composition consisted of Adhesive B adhesive only. The
second consisted of 91% dry weight Adhesive B adhesive
and 9% by weight hollow phenolic microspheres. The
second composition was similar to the first except that
5 the hollow microspheres were glass rather than phenolic.

In each trial, the adhesive composition was heated
to a temperature of about 60°C and gravity fed through a
two-inch pipe into the hopper of the extruder feed unit.
This produced a feed rate and production rate of up to
10 about 32 lb/hr. The temperature of the first barrel
section or feed zone was about 100°C. The temperature
of the second and third barrel sections was also about
100°C. The temperature of the fourth, fifth and sixth
barrel sections was from about 120° to about 150°C. The
15 die temperature was from about 120 to about 140°C and
the melt temperature was about 124°C to about 140°C.
The extruder drive was operated at 70 to 214 rpm. The
vacuum pumps maintained an atmospheric pressure in all
three solvent removal units of approximately 70 torr.
20 Approximately 80% by volume of the solvent was removed
by the first solvent removal unit. Approximately 18% to
19% by volume of the solvent was removed by the second
solvent removal unit and approximately 1% to 2% by
volume of the solvent was removed by the third solvent
25 removal unit. The amount of residual solvent remaining
in all of the compositions was less than about 1%. The

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1 amount of residual free-monomer was found to be less than 0.1%.

EXAMPLE VII

5 A 750 millimeter sheeting die was fitted onto a Berstorff 90 millimeter corotating twin screw extruder. The extruder had seven barrel sections. A feed unit was mounted on the first barrel section. The screw profile of the extruder is shown schematically in FIG. 10.
10 Here, screw element 71 is a return scroll. Screw elements 72 are triple flight conveying elements, specifically Berstorff No. 125-1-3. Screw element 73 is a single flight conveying element, specifically Berstorff No. 125-1-S. Screw elements 74 are double flight conveying elements, specifically Berstorff No. 125-1-2. Screw elements 75 are mixing elements, specifically Berstorff No. ZS-15-25. Screw elements 76 are blisters, specifically Berstorff Blister 88. Screw element 77 is a single flight conveying element, specifically Berstorff No. 100-1-S and screw elements 78 are double flight conveying elements, specifically Berstorff No. 100-1-2. Screw elements 79 and 80 are kneading elements, specifically Berstorff Nos. KS-23-RE and A-KS-23-RE. Finally, screw elements 81 and 82 are
20 double flight conveying elements, specifically Berstorff Nos. A-125-1-2 and A-100-2 respectively.

25 Three solvent removal units were set up involving the fourth, fifth and sixth barrel sections, each having a large vent opening. A two inch diameter duct
30 connected each barrel section of a solvent removal unit with a vacuum pump. A 25 horsepower liquid ring pump used to reduce the atmospheric pressure in all three barrel sections of the solvent removal units. Solvent removed by the solvent removal units was condensed and
35 collected using a condenser.

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1 Trial runs were performed involving three adhesive
compositions. All three compositions had 62% solids,
i.e., 38% by volume solvent which was ethylacetate, etc.
The solids of the first adhesive composition consisted
5 of Adhesive A adhesive only. The solids of the second
adhesive composition consisted of 91% dry weight
Adhesive A, 5.4% by weight Q-Cel 500 hollow glass micro-
spheres and 3.6% by weight Cab-O-Sil M5 fumed silica.
The solids of the third composition consisted of 91% dry
10 weight Adhesive A, 5.4% by weight, Q-Cel 500 hollow
glass microspheres, 3.2% by weight Cab-O-Sil M5 and 0.4%
by weight carbon black Monarch 500.

For the second and third compositions above, the
fillers were pre-blended and fed simultaneously with the
15 adhesive diluted with solvent to the feed unit of the
extruder. The fillers were fed by an Acrison Volumetric
Feeder, and the diluted adhesive was fed by gravity from
two drums.

The temperature of the feed zone (first barrel
20 section) was maintained at 58 to 67°C; the second
section at 84 to 123°C; the third section at 100 to
153°C; the fourth section at 100 to 155°C; the fifth
section at 104 to 148°C; and the sixth section at 144 to
154°C. The die temperature was 107 to 165°C and the
25 melt temperature was 107 to 157°C. The extruder drive
was operated at 100 to 120 rpm. The output ranged from
60 to 150 lbs/hr.

As the adhesive composition was conveyed through
the extruder, approximately 80% of the solvent was
30 removed in the first solvent removal unit, approximately
12% removed in the second solvent removal unit, and 1%
removed in the third solvent removal unit.

Adhesive sheet material was extruded as a sheet
approximately 22 inches wide and 20 to 70 mil thick.
35 The sheet was extruded onto an FEP release film.

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EXAMPLE VIII

A ZE90/90A Berstorff corotating twin screw extruder was set up as described in Example VII with a feed unit at the first barrel section and solvent removal units at the fourth, fifth and sixth barrel sections. A ZE40/40A Berstorff corotating twin screw extruder as described in Example 1 and having approximately 1/10 the capacity of the ZE90/90A extruder was set up at a right angle to the ZE90/90A extruder. The ZE40/40A extruder had a feed unit at the first barrel sections but only two solvent removal units, involving the fourth and fifth barrel sections. The ZE40/40A extruder was joined to the ZE90/90A extruder by an adapter generally as shown in FIG. 5.

A first composition was prepared comprising 53% solids including 91% dry weight Adhesive A adhesive, 5.4% by weight glass microspheres and 3.6% by weight fumed silica. The solvent was a 20:80 mixture of isopropanol and ethyl acetate. A second composition was prepared having 53% solids, the solids comprising Adhesive A adhesive only and the solvent comprising a 20:80 mixture of isopropanol and ethyl acetate.

The first composition was introduced into the ZE90/90A extruder and the second composition was introduced into the ZE40/40A extruder. The ZE90/90A extruder was driven at 140 rpm and the temperature in the second through seventh barrel sections was 102°C, 102°C, 129°C, 131°C, 122°C and 118°C, respectively. The die temperature was 116°C and the melt temperature was 117°C. The ZE40/40A extruder was driven at 197 rpm and the temperatures of the second through seventh barrel sections was 100°C, 100°C, 120°C, 147°C, 186°C and 148°C. The melt temperature was 135°C. The solvent removal units for both extruders was maintained at 120 millibar.

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1 From this arrangement, a double-coated PSA foam
tape was co-extruded at a 200 lb./hr. rate, 20 lb./hr.
from the ZE40/40A extruder. The product had a middle
5 foam layer of the first composition and skin layers of
the second composition. The amount of residual solvents
in the product was less than 0.1%.

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1 WHAT IS CLAIMED IS:

5 1. An adhesive tape having a carrier layer comprising an electron beam-cured polymer matrix, from about 5% to about 70% by volume low density microspheres and at least one pigment in an amount sufficient to impart color to the tape.

10 2. An adhesive tape as claimed in claim 1 wherein the polymer matrix is a pressure sensitive adhesive polymer matrix.

15 3. An adhesive tape as claimed in claim 1, wherein the carrier layer has a thickness of from about 0.25 to about 4.0 mm.

20 4. An adhesive tape as claimed in claim 1, wherein the carrier layer comprises from about 5% to about 45% by volume low density microspheres.

 5. An adhesive tape as claimed in claim 1, wherein the carrier layer comprises from about 10 to about 20% by volume low density microspheres.

25 6. An adhesive tape as claimed in claim 1, wherein the low density microspheres are hollow.

30 7. An adhesive tape as claimed in claim 1, wherein the low density microspheres are made from a material selected from the group consisting of glass, ceramic, polymeric and carbon materials and mixtures thereof.

35 8. An adhesive tape as claimed in claim 7, wherein the low density microspheres are ceramic.

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1 9. An adhesive tape as claimed in claim 7,
wherein the low density microspheres are carbon.

5 10. An adhesive tape as claimed in claim 7,
wherein the low density microspheres are made of a
polymeric material.

10 11. An adhesive tape as claimed in claim 10,
wherein the polymeric material is selected from the
group of phenolic polymers and PVDC copolymers.

15 12. An adhesive tape as claimed in claim 10,
wherein the polymeric material is an inherently tacky,
infusible, pressure sensitive adhesive polymer.

 13. An adhesive tape as claimed in claim 1,
wherein the pigment is carbon black.

20 14. An adhesive tape as claimed in claim 13,
wherein the carbon black is present in an amount of from
about 0.25% to about 5% by weight.

25 15. An adhesive tape as claimed in claim 1,
wherein the carrier layer further comprises fumed silica
in an amount of up to about 10% by weight.

30 16. An adhesive tape as claimed in claim 15,
wherein the fumed silica is present in the carrier layer
in an amount of from about 3 to about 5% by weight.

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1 17. An adhesive tape as claimed in claim 1,
 wherein the carrier layer further comprises up to about
 5% by weight small, rigid high density, solid
5 microspheres having a density of more than about 1.0
 g/cc and an average diameter of less than about 10
 microns.

 18. An adhesive tape as claimed in claim 17,
 wherein the small, rigid high density, solid
10 microspheres have an average diameter of from about 0.1
 to about 5 microns.

 19. An adhesive tape as claimed in claim 1,
 wherein the carrier layer has an elongation of at least
15 about 500 percent and a tensile strength of at least
 about 0.5 to 1.3 megapascals.

 20. An adhesive tape as claimed in claim 1,
 wherein the carrier layer has a storage modulus of at
20 least about 10^4 pascals at 0.01 radians frequency and no
 more than about 5×10^6 pascals at 100 radians frequency
 and a loss modulus of at least 10^4 pascals at 0.01
 radians frequency and no more than about 5×10^6 pascals
 at 100 radians frequency.

25 21. An adhesive tape as claimed in claim 14,
 wherein the carrier layer has a storage modulus of at
 least 4×10^4 pascals at 0.01 radians frequency and no
 more than about 2×10^6 pascals at 100 radians frequency
30 and a loss modulus of at least about 2×10^4 pascals at
 0.01 radians frequency and at least about 2×10^6
 pascals at 100 radians frequency.

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1 22. An adhesive tape as claimed in claim 2,
 wherein the pressure sensitive adhesive polymer matrix
 is acrylic based.

5 23. An adhesive tape as claimed in claim 22,
 wherein the peel adhesion is from about 1300 to about
 3000 newtons/meter.

10 24. An adhesive tape as claimed in claim 2,
 wherein the pressure sensitive adhesive polymer matrix
 is rubber based.

15 25. An adhesive tape as claimed in claim 24,
 wherein the peel adhesion is at least about 3000
 newtons/meter.

 26. An adhesive tape as claimed in claim 25 wherein
 the peel adhesion is at least about 9000 newtons/meter.

20 27. An adhesive tape as claimed in claim 1,
 further comprising a skin layer coated on to at least
 one side of the carrier layer, said skin layer
 comprising an adhesive polymer matrix substantially free
 of low density microspheres.

25 28. An adhesive tape as claimed in claim 27,
 wherein the skin layer has a coating thickness of from
 about 25 to about 125 grams/m².

30 29. An adhesive tape as claimed in claim 1 further
 comprising a skin layer coated onto at least one side of
 the carrier layer, said skin layer comprising a heat
 activatable adhesive substantially free of rigid, low
 density microspheres.

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1 30. A pressure sensitive adhesive tape comprising
a carrier layer having a thickness of from about 0.25 to
about 4.0 millimeters and a skin layer having a coating
thickness of at least about 25 g/m² on at least one side
5 of the carrier layer, said carrier layer comprising a
cross-linked polymer matrix, from about 5% to about 70%
by volume low density microspheres and a colored pigment
in an amount of from about 0.25% to about 5% by weight,
said skin layer comprising a pressure sensitive adhesive
10 polymer matrix substantially free of rigid, low density
microspheres.

15 31. A pressure sensitive adhesive tape as claimed
in claim 30, wherein the carrier layer comprises from
about 10 to about 45% by volume low density
microspheres.

20 32. A pressure sensitive adhesive tape as claimed
in claim 30, wherein the carrier layer further comprises
fumed silica in an amount of up to about 10% by weight.

25 33. A pressure sensitive adhesive tape as claimed
in claim 32, wherein the fumed silica is present in the
carrier layer of an amount of from about 3 to about 5%
by weight.

30 34. A pressure sensitive adhesive tape as claimed
in claim 30, wherein said tape has an elongation of at
least about 300%, a tensile strength of at least 0.5
megapascals, a storage modulus of at least about 10⁴
pascals at 0.01 radians frequency and no more than about
2 x 10⁶ pascals at 100 radians frequency and a loss
modulus of at least 10⁴ pascals at 0.01 radians
frequency and no more than about 2 x 10⁶ pascals at 100
35 radians frequency.

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1 35. A pressure sensitive adhesive tape as claimed
in claim 34, wherein the tape has a storage modulus of
at least 4×10^4 pascals at 0.01 radians frequency and
no more than about 2×10^6 pascals at 100 radians
5 frequency and a loss modulus of at least about 2×10^4
pascals at 0.01 radians frequency and no more than about
 2×10^6 pascals at 100 radians frequency.

10 36. A pressure sensitive adhesive tape as claimed
in claim 30 wherein the polymer matrix is a pressure
sensitive adhesive polymer matrix.

15 37. A pressure sensitive adhesive tape as claimed
in claim 36, wherein the pressure sensitive adhesive
polymer matrix is acrylic based and the peel adhesion is
from about 1300 to about 3000 newtons/meter.

20 38. A pressure sensitive adhesive tape as claimed
in claim 36, wherein the pressure sensitive adhesive
polymer matrix is rubber based and the peel adhesion is
at least about 3000 newtons/meter.

25 39. A pressure sensitive adhesive tape as claimed
in claim 38 wherein the peel adhesion is at least about
9000 newtons/meter.

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1 40. A pressure sensitive adhesive tape comprising
a carrier layer having a thickness of from about 0.25 to
about 4.0 millimeters and a skin layer having a coating
thickness of at least about 25 g/m² on each side of the
5 carrier layer, said carrier layer comprising an electron
beam-cured pressure sensitive adhesive polymer matrix,
from about 10% to about 70% by volume low density
microspheres selected from the group consisting of
glass, ceramic, polymeric and carbon materials and
10 mixtures thereof, a pigment in an amount of from about
0.25% to about 5% by weight; and wherein said skin layer
comprises a pressure sensitive adhesive matrix
substantially free of rigid, low density microspheres.

15 41. A pressure sensitive adhesive tape as claimed
in claim 37, wherein the low density microspheres are
ceramic.

20 42. A pressure sensitive adhesive tape as claimed
in claim 37, wherein the low density microspheres are
carbon.

25 43. A pressure sensitive adhesive tape as claimed
in claim 40, wherein the low density microspheres are
made of a polymeric material.

30 44. A pressure sensitive adhesive tape as claimed
in claim 43, wherein the polymeric material is selected
from the group of phenolic polymers and PVDC copolymers.

35 45. A pressure sensitive adhesive tape as claimed
in claim 43, wherein the polymeric material is an
inherently tacky, infusible, pressure sensitive adhesive
polymer.

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1 46. A process for producing curable adhesive material, said process comprising:

 introducing into an extruder an adhesive composition comprising a curable polymer matrix and
5 solvent for the adhesive matrix.

 conveying the adhesive composition through the extruder;

 reducing the atmospheric pressure in at least one section of the extruder sufficiently to evaporate at
10 least a portion of the solvent in the adhesive composition conveyed through that section;

 removing the evaporated solvent from the extruder; and

 extruding a solvent-reduced curable
15 adhesive material through a die.

 47. A process as claimed in claim 46 wherein the solvent is present in the composition in an amount of
20 from about 20% to about 60% by volume of the composition;

 48. A process as claimed in claim 46, wherein substantially all of the solvent is removed from the
25 adhesive composition so that a solvent-free adhesive material is produced.

 49. A process as claimed in claim 46 wherein the extruded adhesive material has residual free-monomer
30 concentration of less than about 0.1% by weight.

 50. A process as claimed in claim 46 wherein the extruded adhesive material is essentially free of
 bubbles.

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1 51. A process as claimed in claim 46, wherein the
adhesive composition is heated to a temperature of from
about 100°C to about 160°C as it is conveyed through the
extruder.

5 52. A process as claimed in claim 46, wherein the
atmospheric pressure is reduced in at least two sections
of the extruder sufficiently to evaporate at least a
portion of the solvent in the adhesive composition
10 conveyed through each section.

 53. A process as claimed in claim 46 further
comprising electron beam curing the extruded adhesive
material.

15 54. A process for producing curable pressure
sensitive adhesive material, said process comprising:
 providing a twin screw extruder having at
least one upstream feed unit and at least one downstream
20 solvent removal unit, said solvent removal unit
comprising a barrel section having a large vent opening,
a vacuum pump, and a duct surrounding the vent opening
and extending from the barrel section to the vacuum pump
whereby the vacuum pump can be activated to reduce the
25 atmospheric pressure within the barrel section, vent
opening and duct;

 introducing into the feed unit of the extruder
an adhesive composition comprising a curable adhesive
polymer matrix and a solvent for the adhesive polymer
30 matrix, said solvent being present in an amount of from
about 20% to about 60% by volume of the adhesive
composition;

 heating the composition and conveying the
composition through the extruder;

35 activating the vacuum pump to reduce the

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1 atmospheric pressure in the barrel section of the
solvent removal unit sufficiently to evaporate at least
a portion of the solvent in the adhesive composition
conveyed through that section; and

5 extruding curable adhesive material through
die.

55. A process as claimed in claim 54, wherein the
extruder comprises at least two solvent removal units.

10 56. A process as claimed in claim 54, wherein the
extruded curable adhesive material is solvent-free.

15 57. A process as claimed in claim 54 wherein the
residual free-monomer concentration of the extruded
curable adhesive material is less than about 0.1% by
weight.

20 58. A process as claimed in claim 54, wherein the
adhesive composition is heated to a temperature of from
about 100°C to about 160°C as it is conveyed through the
extruder and the pressure in the solvent removal units
is reduced from about 150 to about 100 torr.

25 59. A process as claimed in claim 54 wherein the
adhesive composition comprises at least one filler and
wherein the filler is introduced into the extruder
separately from the curable adhesive polymer matrix and
solvent.

30 60. A process as claimed in claim 59 wherein the
filler is selected from the group consisting of low
density microspheres, pigments, fumed silica and
mixtures thereof.

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1 61. A process as claimed in claim 59 wherein the
extruder comprises a downstream feed unit and the
curable pressure sensitive adhesive polymer matrix and
solvent are introduced into the upstream feed unit and
5 the filler is introduced in the downstream feed unit.

10 62. A process as claimed in claim 61 wherein the
downstream feed unit is located downstream from the
solvent removal units.

15 63. A process as claimed in claim 59 wherein the
upstream feed unit comprises a first feed port on the
lower side of the extruder and a second feed port on the
upper side of the extruder and wherein the curable
adhesive polymer matrix and solvent are introduced into
the extruder through the first feed port and the filler
is introduced through the second feed port.

20 64. A process as claimed in claim 54 further
comprising:

25 providing a second twin screw extruder having
at least one upstream second feed unit and at least one
downstream second solvent removal unit, said second
solvent removal unit comprising a barrel section having
a large vent opening, a vacuum pump, and a duct
surrounding the vent opening and extending from the
barrel section to the vacuum pump whereby the vacuum
pump can be activated to reduce the atmospheric pressure
within the barrel section, vent opening and duct;

30 connecting the first and second extruders so
that product from both extruders are extruded through a
single die;

35 introducing into the feed unit of the second
extruder a second adhesive composition comprising a
second curable adhesive polymer matrix and solvent for

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1 the second adhesive polymer matrix, said solvent being
present in an amount of from about 20% to about 60% by
volume of the second adhesive composition;

heating the second composition and conveying
5 the second composition through the second extruder;

activating the vacuum pump of the second
solvent removal unit to reduce the atmospheric pressure
in the barrel section of the second solvent removal unit
sufficiently to evaporate at least a portion of the
10 solvent in the second adhesive composition conveyed
through that section; and

co-extruding curable adhesive having at least
one layer of the first adhesive composition with reduced
solvent content and at least one layer of the second
15 adhesive composition having reduced solvent content.

65. A process as claimed in claim 64 wherein the
extruded curable adhesive is solvent free.

20 66. A process as claimed in claim 64 wherein the
extruded curable adhesive has a residual monomer
concentration of less than about 0.1% by weight.

25 67. A process as claimed in claim 64 wherein the
curable adhesive is extruded as a sheet comprising a
middle layer of the first adhesive composition with
reduced solvent content and two skin layers of the
second adhesive composition with reduced solvent
content.

30 68. A process as claimed in claim 67 wherein the
first adhesive composition comprises low density
microspheres and the second adhesive composition is
essentially free of rigid, low density microspheres.
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1 69. A process as claimed in claim 64 further comprising electron beam curing the extruded curable adhesive.

5 70. A process as claimed in claim 54 wherein the die is a sheet die and comprises spaced-apart upper and lower plates forming a chamber therebetween and wherein at least one of the upper and lower plates comprises one or more of holes extending from the outer surface of the
10 plate into the chamber, said sheet die further comprising a manifold mounted on the exterior surface of at least one of the upper and lower plates in surrounding relation to the holes, and wherein the process further comprises:

15 introducing into the manifold, a second adhesive composition; and

 co-extruding a sheet of curable adhesive material comprising a carrier layer of the first adhesive composition with reduced solvent content and
20 strips of the second adhesive composition at the surface of the carrier layer.

 71. A process as claimed in claim 70 wherein the strips are broken, forming patches of the second
25 adhesive composition at the surface of the carrier layer.

 72. A process as claimed in claim 54 wherein at least a portion of the screws of the twin screw extruder
30 are intermeshing.

 73. A process as claimed in claim 54 wherein at least a portion of the screws of the twin screw extruder are tangential.

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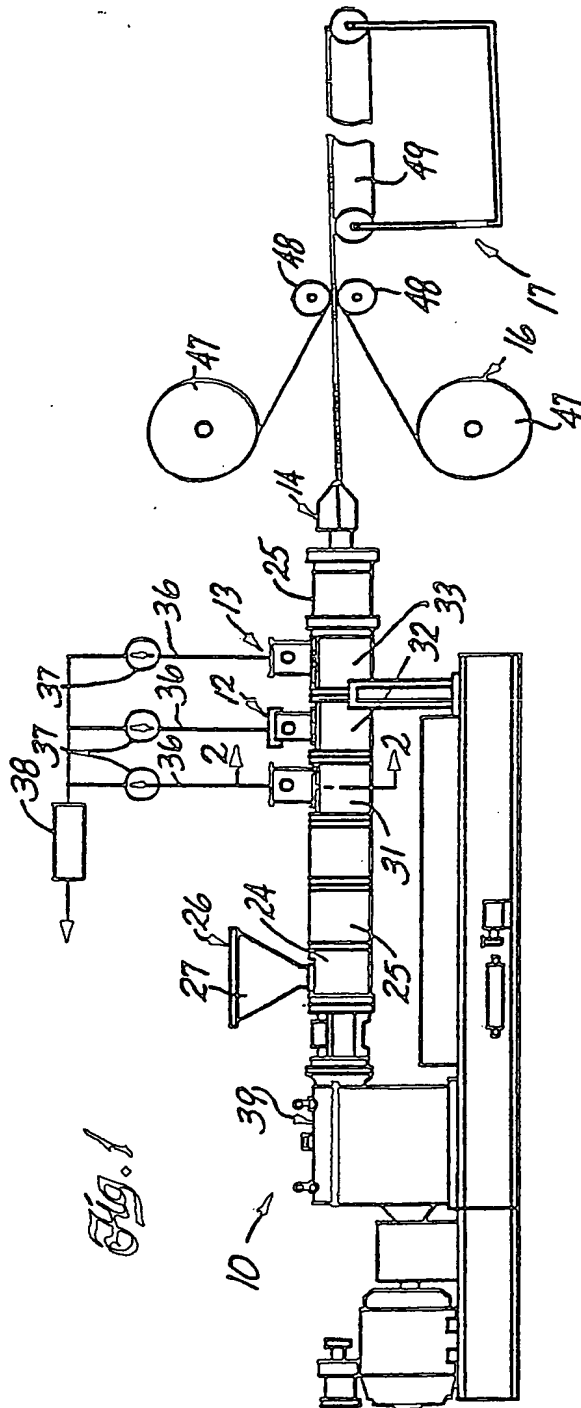


Fig. 2

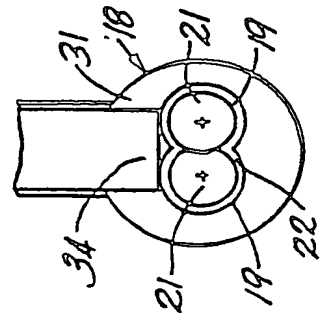


Fig. 3

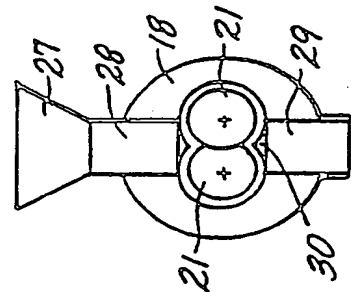


Fig. 4

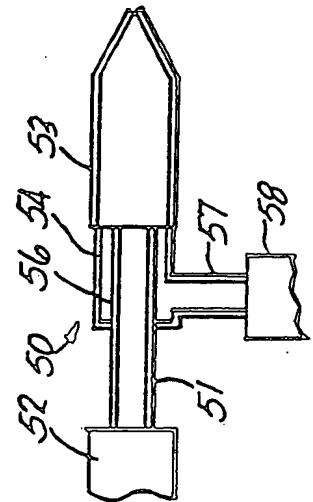


Fig. 4

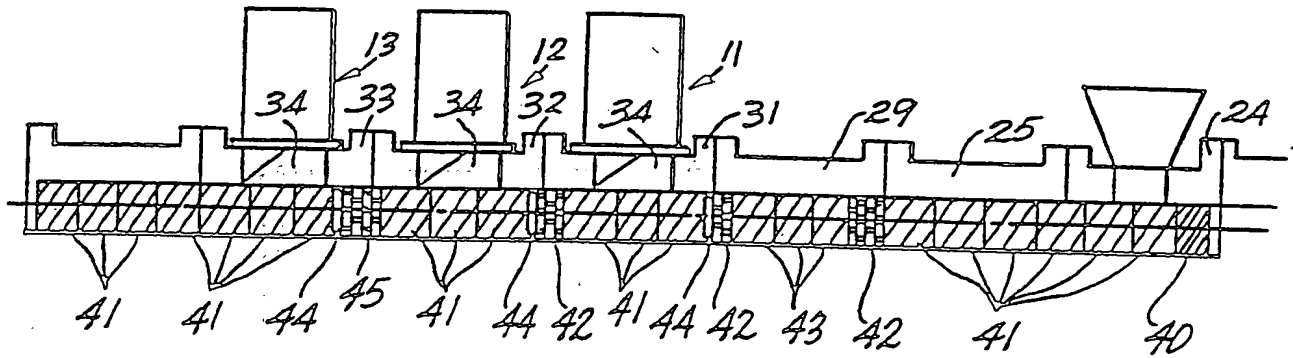


Fig. 9

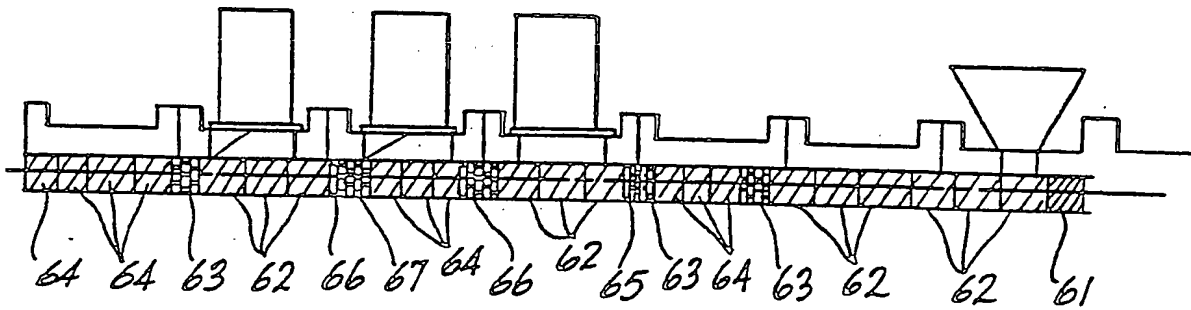
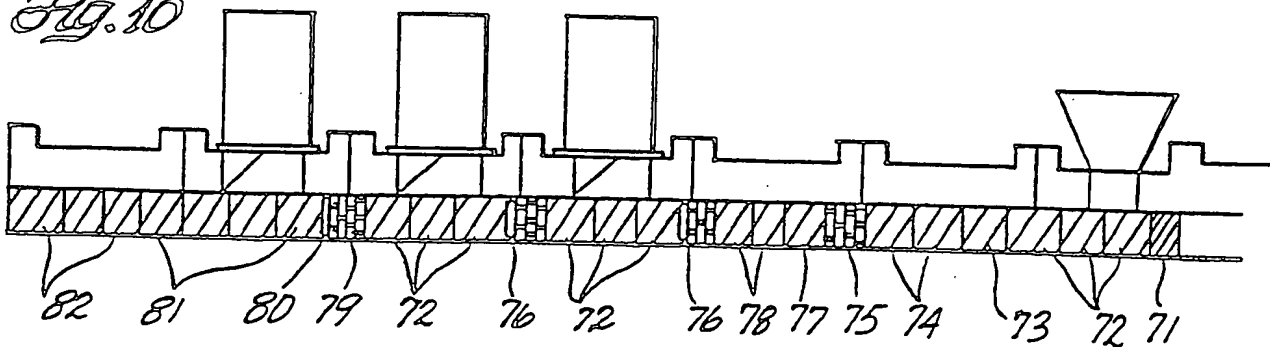
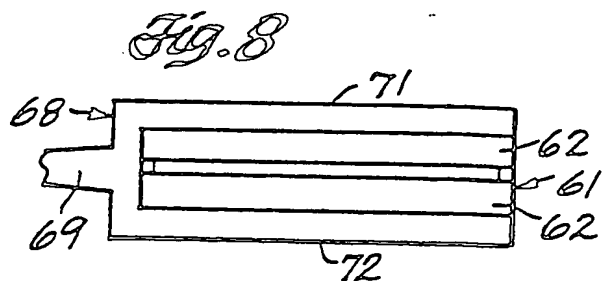
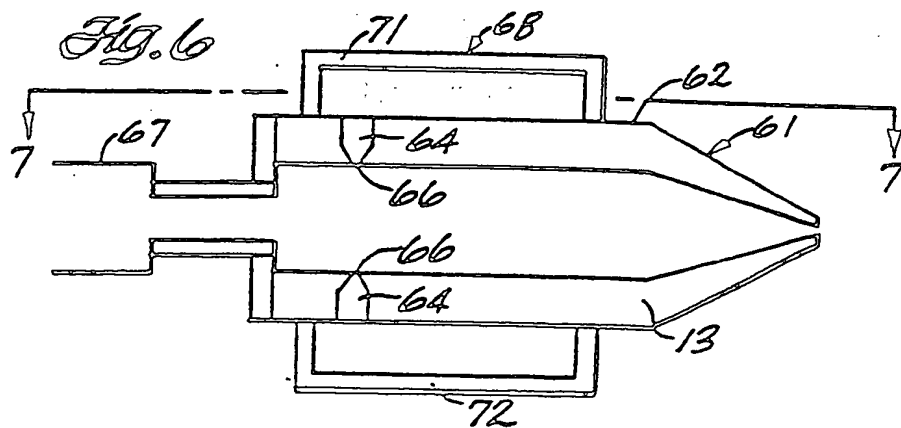
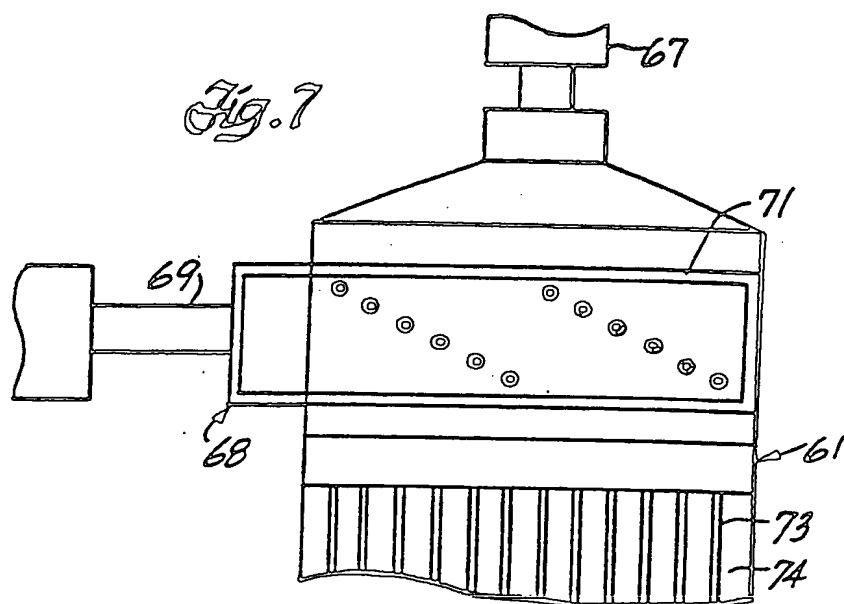


Fig. 10





INTERNATIONAL SEARCH REPORT

International Application No. PCT/US88/021.93

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) ⁶

According to International Patent Classification (IPC) or to both National Classification and IPC

INT. CL. 4 B32B 5/16

U.S. CL. 265/101, 171; 425/203; 428/313.3, 325, 327

II. FIELDS SEARCHED

Minimum Documentation Searched ⁷

Classification System

Classification Symbols

U.S.

264/101, 171 425/203;
428/313.3, 313.5, 313.9, 323, 325, 327

Documentation Searched other than Minimum Documentation
to the extent that such Documents are included in the Fields Searched ⁸

III. DOCUMENTS CONSIDERED TO BE RELEVANT ⁹

Category [*]	Citation of Document, ¹¹ with indication, where appropriate, of the relevant passages ¹²	Relevant to Claim No. ¹³
Y, P	US, A, 4,748,061 (VESLEY) 31 MAY 1988 ENTIRE DOCUMENT	1-73
Y	US, A, 4,223,067 (LEVINS) 16 SEPTEMBER 1980. COLUMN 3, LINES 16-21.	1-73
Y	US, A, 4,098,945 (OEHRKE) 04 JULY 1978. ENTIRE DISCLOSURE.	1-73
Y	US, A, 4,065,532 (WILD) 27 DECEMBER 1977	46-73
Y	US, A, 3,985,348 (SKIDMORE) 12 OCTOBER 1976. ENTIRE COLUMNS 2 AND 3.	64-69
Y	US, A, 3,565,737 (LEFEVRE) 23 FEBRUARY 1971	70-73

^{*} Special categories of cited documents: ¹⁰

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

IV. CERTIFICATION

Date of the Actual Completion of the International Search

26 SEPTEMBER 1988

Date of Mailing of this International Search Report

22 NOV 1988

International Searching Authority

ISA/US

Signature of Authorized Officer

W. J. VANBALEN
W. J. Van Balen